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**AN INVESTIGATION OF AEROSOL MEASUREMENTS FROM THE
HALOGEN OCCULTATION EXPERIMENT: VALIDATION, SIZE
DISTRIBUTIONS, COMPOSITION, AND RELATION TO OTHER
CHEMICAL SPECIES**

to: The University of Wyoming

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1. Introduction

The efforts envisioned within the original proposal (accepted February 1994) and the extension of this proposal (accepted February 1997) included measurement validations, the retrieval of aerosol size distributions and distribution moments, aerosol correction studies, and investigations of polar stratospheric clouds. This report provides a summary of the efforts completed under funding for this grant, which expired June 30, 1998.

A majority of the results from this grant have been published in the refereed literature and/or constitute portions of Mark Hervig's Ph.D. dissertation [*Hervig et al.*, 1995; *Hervig et al.*, 1996a; *Hervig et al.*, 1996b; *Hervig*, 1997; *Hervig et al.*, 1997; *Hervig et al.*, 1998; *Hervig and Deshler*, 1998]. The principal results from this grant are discussed below, and reprints of journal articles are attached.

2. Completed Studies

2.1. Aerosol Corrections

The HALOE aerosol correction scheme was finalized and published. Understanding and removing the aerosol extinction is essential for obtaining accurate retrievals from the HALOE radiometer channels of NO₂, H₂O and O₃ in the lower stratosphere since these measurements are severely affected by contaminant aerosol absorption. If ignored, aerosol absorption in the radiometer measurements is interpreted as additional absorption by the target gas, resulting in anomalously large mixing ratios. To correct the radiometer measurements for aerosol effects, a retrieved aerosol extinction profile is extrapolated to the radiometer wavelengths and then included as continuum attenuation. The sensitivity of the extrapolations to size distribution and composition is small for certain wavelength combinations, reducing the correction uncertainty. The aerosol corrections extend the usable range of profiles retrieved from the radiometer channels to the tropopause with results that agree well with correlative measurements. In situations of heavy aerosol loading, errors due to aerosol in the retrieved mixing ratios are reduced to values of about 15, 25, and 60% in H₂O, O₃, and NO₂, respectively, levels that are much less than the correction magnitude [*Hervig et al.*, 1995].

2.2. Aerosol Measurement Validation

A validation of the HALOE aerosol measurements was completed and published. This work was based on uncertainty analysis, internal validations, comparisons with theory, and comparisons with independent measurements. Monte Carlo calculations using accepted values of random and systematic errors determine typical measurement uncertainties of less than 15% for pressures from 100 to 10 mb. Comparisons of coincident HALOE sunrise and sunset observations indicate systematic differences (sunrise > sunset) for pressures less than 10 mb. Random sunrise-sunset

differences, taken as an upper limit of the measurement precision, are generally from 10 to $\sim 30\%$ for pressures from 100 to 10 mb. Measured extinction ratios were compared with ratios determined from theory. These comparisons show that the measurements are consistent with theory at pressures from 100 to 10 mb, depending on channels, latitude, and season. HALOE extinctions were compared with extinctions calculated from balloonborne particle counter measurements. The results show random differences from 30 to 50% for pressures from 100 to 10 mb and systematic differences (HALOE > particle counters) for pressures less than 40 mb. The results indicate that the HALOE 2.80 μm aerosol measurements are much less reliable than the other four measurements [Hervig *et al.*, 1996a].

2.3. Temperature Measurement Validation

A validation of the HALOE temperature measurements was completed and published. Temperatures are retrieved from HALOE transmission measurements in the in the 2.80- μm CO_2 band for levels above where aerosol affects the signals (35 km) to altitudes where the signal-to-noise decreases to unity (~ 85 km). At altitudes from 45 to 35 km the profile undergoes a gradual transition from retrieved to National Meteorological Center (NMC) temperatures and below 35 km the profile is strictly from the NMC. This validation covered the uncertainty analysis, internal validations, and comparisons with independent measurements. Monte Carlo calculations using all known random and systematic errors determine typical measurement uncertainties of 5 K for altitudes below 80 km. Comparisons of coincident HALOE sunrise and sunset measurements are an indicator of the upper limit of measurement uncertainty. The sunrise-sunset comparisons have random and systematic differences which are less than 10 K for altitudes below 80 km. Comparisons of HALOE to lidar and rocket measurements typically have random differences of ~ 5 K for altitudes below 65 km. The mean differences for the correlative comparisons indicate that HALOE temperatures have a cold bias (2 to 5 K) in the upper stratosphere and stratopause [Hervig *et al.*, 1996b].

2.3. Polar Stratospheric Cloud Studies

Investigations were conducted to identify the formation and composition of polar stratospheric clouds (PSCs). These efforts used the HALOE aerosol and water vapor measurements in combination with model simulations of PSC growth. An important result of this study was quantifying a mechanism for late spring PSC formation under dehydrated and denitrified polar vortex conditions. HALOE observations over the eastern Weddell Sea during early October 1993 show factor of ~ 30 increases in aerosol volume coincident with water vapor enhancements of ~ 3 ppmv, suggesting that vapor intrusions were an important factor in determining PSC growth. Comparing the measurements with model calculations confirms this, and demonstrates that the

HALOE aerosol and water vapor measurements are quantitatively consistent with thermodynamic predictions of PSC particle volumes [Hervig *et al.*, 1997].

2.4. Aerosol Size Distribution Retrievals

The ability to reliably retrieve the aerosol size distribution (in addition to the surface area and volume) satisfies important needs for investigations of the Earth's radiative and chemical balance. A technique was developed to use multi-wavelength aerosol extinction measurements from HALOE to determine the size distribution of stratospheric sulfate aerosols. Although the HALOE extinction spectrum alone cannot be used to reliably infer the aerosol size distribution (except when the aerosol population contains particles larger than about 0.5 μm), the inverse problem becomes highly defined when the effective radius is known. Using theoretical relationships derived from in situ aerosol measurements, it was found that the effective radius can be determined from the HALOE 2.45 μm extinction with uncertainties of about $\pm 15\%$. Using extinction ratios with the effective radii determined from the HALOE extinctions, unimodal lognormal size distributions were obtained through explicit solutions. The HALOE size distributions are in excellent agreement with coincident in situ aerosol measurements.

This work has been published [Hervig *et al.*, 1998], and the algorithms developed for this work were delivered to the HALOE team at NASA Langley where they were used to retrieve size distribution profiles from the entire HALOE data set. These data were then made available to the public on the World Wide Web under the HALOE home page.

2.5. Direct Inference of Aerosol Surface Area and Volume

Relationships were developed for converting HALOE measurements of stratospheric aerosols to surface area and volume densities. The relationships were derived using an extensive record of in situ aerosol size distribution measurements made in sulfate aerosols and polar stratospheric clouds (PSCs). Conversions were derived for sulfate aerosols using the appropriate refractive indices, and for PSCs using the refractive indices for either nitric acid trihydrate or liquid ternary $\text{H}_2\text{SO}_4\text{--H}_2\text{O--HNO}_3$ aerosols. Coincident measurements from HALOE and in situ particle counters are in good agreement at middle latitudes, for surface area and volume densities. This method allows results to be easily obtained by HALOE data users, for both sulfate aerosols and PSCs. Additionally, the surface areas and volumes obtained from this method can be compared to values obtained from the size distribution retrievals (described above), as a consistency check of the two methods [Hervig and Deshler, 1998].

3. References

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